VĚDECKÉ SPISY VYSOKÉHO UČENÍ TECHNICKÉHO V BRNĚ

Edice Habilitační a inaugurační spisy, sv. 701
ISSN 1213-418X

Vlastimil Křápek

NANOSTRUCTURES FOR QUANTUM INFORMATION PROCESSING

BRNO UNIVERSITY OF TECHNOLOGY
FACULTY OF MECHANICAL ENGINEERING
INSTITUTE OF PHYSICAL ENGINEERING

Mgr. Vlastimil Křápek, Ph.D.

NANOSTRUCTURES FOR QUANTUM INFORMATION PROCESSING

NANOSTRUKTURY PRO KVANTOVÉ ZPRACOVÁNÍ INFORMACE

SHORT VERSION OF HABILITATION THESIS
IN APPLIED PHYSICS

VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ
NAKLADATELSTVÍ VUTIUM

BRNO 2021

Key words

Nanostructures, quantum dots, exciton, fine structure splitting, quantum information processing, quantum entanglement, polarization entanglement, entangled photon pairs, k·p theory, configuration interaction, plasmonic antennas, Purcell effect, electron energy loss spectroscopy.

Klíčová slova

Nanostruktury, kvantové tečky, exciton, rozštěpení jemné struktury, kvantové zpracování informace, kvantové propletení, polarizační propletení, propletené dvojice fotonů, teorie k·p, konfigurační interakce, plazmonické antény, Purcellův jev, spektroskopie energiových ztrát elektronů.

Místo uložení práce

Habilitační práce je uložena v areálové knihovně FSI VUT v Brně.

ISBN 978-80-214-5985-4
ISSN 1213-418X

Contents

Author

Vlastimil Křápek's primary area of interest is a theoretical and numeric modeling of various nanoscale systems, including semiconductor quantum dots, quantum dot molecules, metallic nanoparticles, and their complexes. He studies the interrelation between the structural, electronic, and optical properties of quantum dots as well as their tunability by external effects (electric, magnetic, and strain field). In collaboration with experimentalists he pursues technologically important goals to push the emission of InAs quantum dots to the infrared telecommunication wavelengths or to lower the excitonic fine structure splitting below the natural linewidth. Recently he turned his attention to the metallic particles hosting localized surface plasmon resonances. He is involved in the modeling of electron energy loss spectroscopy of plasmonic nanoparticles and interested in coupling of the plasmonic particles with quantum dots, focusing on the enhancement of their photoluminescence rate (Purcell effect). He led three national-level research projects and participated in many others. He coauthored 52 scientific papers to which over 900 references were made, and achieved Hirsch index of 15. His teaching activities include lectures (Oscillations, waves, and optics; Fundamentals of nanosciences) and seminars (Fundamentals of physics), supervision of students (1 bachelor, 4 master, 2 PhD as a supervisor specialist), and development of learning resources.

Dedication

To all people who are filling
Plenty of Room at the Bottom.

Acknowledgment

My sincere appreciation goes to my colleagues and collaborators who created an inspiring research environment in Brno, Berlin, Dresden, Prague and elsewhere. Nature is acknowledged as the bottomless source of puzzles, and Physics as the ultimate tool that, with only a bit of ingenuity and imagination, allows to discover.

1 Introduction

1.1 The splendors and miseries of nanostructures

The central point of this thesis are *nanostructures* – objects with the dimensions on the scale of nanometers (or up to hundreds of nanometers). The field of nanostructures is tremendously huge and I will cover only a small part here, based on my experience and preferences: Quantum dots and plasmonic antennas.

One thing all nanostructures have in common is that they are small. Miniaturization brings many advantages. It makes devices cheaper, portable, consuming less supplies or integrable into more complex functional units, all this while preserving the functionality. Many examples of successful miniaturization include cell phones, computers, transistors, or clockworks. Although it is common to consider the miniaturization a very modern trend, the last case shows that it is rather old, dating back to medieval times or even before.

In the aforementioned examples the smaller dimensions have not brought in the new functionality. Novel physics emerges when the dimensions of the nanoparticle become comparable with a characteristic length of some physical process, particle, or interaction. As the first example we will consider light. Its characteristic dimension is its wavelength (spatial periodicity) and for visible light in vacuum it ranges approximately from 400 nm to 750 nm. In the homogeneous medium, the vacuum wave length has to be divided by the refractive index, usually larger than one. When the medium is structured on the scale comparable with the light wave length, light behaves very differently than in the free space. A prominent example is a *photonic crystal* [1, 2, 3] formed by a periodic arrangement of building blocks with a contrast in the refractive index, such as a dielectric slab with drilled holes. A photonic band structure is formed in photonic crystals similar to the electron energy band structure in ordinary crystals, including the photonic band gaps – ranges of photon energies for which the absorption or emission of light (including spontaneous emission) is not allowed. On the other hand, for different photon energies the density of photon states can be considerably enhanced comparing to the free space case (described by Planck's law). In consequence, the emission of light can be enhanced. This effect was discovered by Edward Mills Purcell [4] (hence the name Purcell's effect) and is employed to improve the properties of light emitters [5, 6, 7]. *Photonic metamaterials* are periodic structures similar to photonic crystals but with subwavelength periodicity [8, 9, 10]. Building blocks represent artificial atoms, metaatoms, with engineerable properties. Metamaterial is perceived as a homogeneous environment by light as long as the wave length of light is much larger than the metaatom dimensions. It is thus well described by the local frequency-dependent electric permittivity and magnetic permeability. However, tremendous variability of metaatoms in comparison with atoms allows for extraordinary properties, a prominent example of which is a negative refractive index [8, 9, 11]. Metamaterial are also used for cloaking – making object invisible by allowing light to travel around the object instead of being scattered or absorbed [10, 12]. *Cavity quantum electrodynamics* [13, 14], another branch of photonics, focuses on light confined in a photonic cavity and interacting with a matter (an optical emitter). When the light is squeezed to sufficiently small volume (i.e., the cavity mode volume is comparable with λ^3 where λ denotes the wave length) and kept in the cavity for sufficiently long time (i.e., the cavity quality factor Q has to be large enough), then a coherent superposition of the light and matter states is formed, resulting in Rabi oscillations [15].

Plasmonic antennas are yet another kind of nanostructures confining light. In contrast to dielectric photonic structures discussed in the previous paragraph, plasmonic particles are metallic. The interface between the metal and dielectric hosts surface plasmon polaritons, which are electromagnetic waves coupled to the oscillations of free electron gas in the metal. The waves are evanescent:

they decay exponentially into both metal and dielectric. They can propagate along the interface. However, when the interface is enclosing the particle and is thus of finite size, standing waves are formed instead – localized surface plasmons. Due to their evanescent nature they can be confined on a subwavelength scale. Plasmonic modes have therefore a very small volume but their quality factor is lower than in the dielectric photonic structures due to ohmic losses in the metal.

Electrons experience more complex interaction with the environment than light. A discrete character of electric charge can be demonstrated in a device known as single electron transistor (SET). This is composed of a central capacitor of capacitance C connected via tunnel junctions to conducting electrodes (source and drain). With a large enough capacity, charging the capacitor with an extra electron costs almost no energy and the current flows through the transistor depending on the bias voltage and the resistivity of the device (mostly related to the tunnel junctions). However, when the charging energy of the capacitor, $E_C = e^2/2C$ (e denotes the elementary charge) becomes comparable with the thermal energy $E_T = k_B T$ (k_B is the Boltzmann constant and T is the temperature), the current is blocked for sufficiently small bias voltage – the effect is known as Coulomb blockade.The current can be restored, though, either by increasing the bias voltage above a certain threshold, or by gating the central capacitor and applying a gate voltage that compensates for the charging energy. Considering a spherical capacitor ($C = 4\pi\varepsilon R$), a characteristic dimension (the radius of the capacitor R) reads

$$R = \frac{e^2}{8\pi\varepsilon k_B T}$$

which, for $T = 300$ K and the dielectric constant ε of vacuum, takes the value $R = 39$ nm.

Electrons can also be confined in a way similar to light. Here, the characteristic length is de-Broglie wave length, reading $\lambda_{dB} = h/p$ (h is Planck constant and p is the momentum of the electron). For a non-relativistic electron with the kinetic energy E it reads $\lambda_{dB} = h/\sqrt{2mE}$. Inserting the thermal energy at 300 K and the effective mass of 0.067 of free electron mass m_e (corresponding to the edge of the conduction band in GaAs), one gets the de Broglie wave length of 30 nm. The nanoobjects of similar or smaller dimensions have the electron energy structure significantly different from extended objects which results into their non-classical properties.

1.2 Fabrication

The progress in the field of nanostructures is intimately related to the development of ingenious fabrication techniques. Traditionally, they fall into top-down and bottom-up approaches. In a *top-down* approach, the starting point is a bulk material that is processed (etched, milled, sputtered off, etc.) into a desired size and shape. Top-down techniques are usually deterministic and versatile, but on the other hand slow and limited to rather large objects. In a *bottom-up* approach, atoms, molecules, or other complexes are used as building blocks for more complex structures, put together e.g. by chemical reactions or by nanomanipulation. Bottom-up approaches are rather stochastic, difficult to control, and not so versatile, but they are fast, often scalable and suitable for mass production, and allow for smaller nanostructures than top-down approaches. We note that modern fabrication techniques are often composed of several steps combining both approaches. In the following, we present some of the basic fabrication techniques.

Electron beam lithography (EBL) is a technique capable to produce planar structures of arbitrary shape with the lateral resolution better than 10 nm. A substrate is first coated with an electron-sensitive resist. A focused electron beam is used to draw the desired structure, locally modifying the properties of the resist. Next, the development follows in which the exposed (positive lithography) or unexposed (negative lithography) resist is removed. Finally, a layer of the desired material is deposited and the remaining resist is removed.

Focused ion beam milling (FIB) is a technique in which the desired structure is drawn with the focused beam of ions (most often Ga or Xe) directly into the thin layer of the target material. Contrary to electrons, massive ions are capable to sputter-off the target material so there is no need for the resist and development.

Nanoimprint lithography uses a solid stamp to structure the soft resist (by a mechanical pressure), which is then processed as in the case of EBL. In comparison with the previously mentioned lithographic techniques, nanoimprint lithography is low-cost and high-throughput technique suitable for industrial applications.

Nanomanipulation was enabled by the development of scanning probe microscopy. The objects (individual ions or larger particles) can be attached to the tip of the probe, dragged over the substrate, and detached at the desired position. The technique has been used e.g. to arrange 35 xenon atoms on nickel substrate to spell out the letters IBM or to place the plasmonic particles to the vicinity of self-organized quantum dots [16].

Chemical synthesis encompasses a broad class of chemical methods, in which nanostructures are produced through chemical reactions. The nanostructures prepared via chemical synthesis are usually of high quality (as no dirty or damaging processes are involved such as the resist deposition in EBL or the mechanical treatment in FIB). On the other hand, there is only a limited control over the process and its products, and also a limited versatility. Using chemical synthesis, it is possible to produce e.g. single crystal gold platelets for applications in plasmonics [17, 18], colloidal quantum dots [19], molecular nanomagnets [20, 21], etc.

Epitaxy refers to the ordered layer-by-layer growth of a crystalline film on a crystalline substrate. It produces high-quality thin films. The growth rate is rather small, between tenths of nanometer and units of micrometer per minute. In combination with using metallic nanoparticles as seeds (vapor-liquid-solid mechanism [22]), one-dimensional epitaxial growth can be realized producing nanowires. Quantum dots can be produced as well, either by self-assembly of the lattice-mismatched heterostructures, or by droplet epitaxy (see Sec. 1.4.1).

1.3 Characterization

The structural characterization of nanostructures relies on two main approaches: scanning probe microscopy and electron microscopy.

Scanning probe microscopy (SPM) is a class of methods in which the surface of the sample is scanned with a probe in a form of a sharp tip. Piezoelectric actuators allows for a precise control of the tip position. The methods differ by the physical quantity detected by the tip. *Atomic force microscopy* (AFM) relies on short-range mechanical force between the probe and the sample. It allows to image the surface profile with the sub-nanometer vertical resolution and can also be used for nanomanipulation. *Scanning tunneling microscopy* measures the tunneling current between the conductive tip and the sample. It is restricted to conductive samples. Due to the exponential dependence of the tunneling current on the tip-sample distance it provides a superb spatial resolution of 10 pm vertically and 100 pm horizontally [23]. Further, by varying the bias voltage the method enables to retrieve the energy-dependent local density of states. In *Scanning near-field optical microscopy*, the probe has a form of a tapered optical fibre that collects the near electromagnetic field (i.e., evanescent waves related e.g. to the quantum emitters or surface plasmons) present at the surface of the sample. The lateral resolution down to 20 nm has been demonstrated [23], well below the far-field diffraction limit.

Electron microscopy employs a beam of electrons to probe a sample. Owing to their short de Broglie wave length, electrons enable considerably better spatial resolution (several tens pm) than light (several hundreds of nm). The microscope is composed of an electron source (electron

gun), a set of lenses, a sample stage, and a detector (usually a CCD camera), all mounted in evacuated electron column. There are two principal configurations of the electron microscope. In a *scanning electron microscope* (SEM), the focused electron beam is scanned over the sample and a reflected signal (secondary electrons, or possibly back-scattered electrons, X-rays, or light due to cathodoluminescence) is recorded. In a *transmission electron microscope* (TEM) a broad electron beam is transmitted through a sample. In addition to the unscattered electrons, the signal is composed of scattered electrons and cathodoluminescence. TEM requires thin samples (several tens of nanometers for the highest spatial resolution) and high-energy electrons that penetrate the sample (100–300 keV compared to less than 30 keV used in SEM). The scanning mode is also available for TEM, which is then known as scanning TEM (STEM).

Optical properties of nanoparticles and to some extent their electronic and quasiparticle structure are most conveniently studied by optical methods. Most prominent of them in the field of nanostructures are optical transmission and reflection spectroscopy and luminescence.

Optical transmission and reflection spectrometry is a quantitative method allowing to determine both the energy and the strength of various transitions (electronic, phononic, etc.) in a matter. White light from a source is either transmitted through or reflected from a sample, monochromatized or dispersed, and detected.

For the interaction of a matter with light, usually the Fermi golden rule and the electric dipole approximation are appropriate. The absorption transition rate (number of photons being absorbed per second) for the transition from the initial single electron state i to the final single electron state f then reads

$$W_{fi} = \frac{2\pi}{\hbar}\left(\frac{eE_0}{m_e\omega}\right)^2 |\langle f|\mathbf{e}\cdot\hat{\mathbf{p}}|i\rangle|^2\delta(E_f - E_i - \hbar\omega),$$

where $\hat{\mathbf{p}} = -i\hbar\nabla$ is the momentum operator, E_0, $\mathbf{e}$, and ω denote the amplitude, polarization vector, and frequency of the electric field, $E_{i,f}$ are the energies of respective states, and e, m_e, and $\hbar$ are the charge and mass of an electron, and reduced Planck constant. For more detailed discussion we refer to the standard textbooks, e.g., [24, 25]. Similar relations can be derived for other transitions related to the absorption of light (e.g. phononic or excitonic).

For a sample that is (at least piecewise) homogeneous it is customary to introduce the complex conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$, whose real part reads

$$\sigma_1(\omega) = \frac{\pi e^2}{m_e^2\omega}\frac{2}{\Omega}\sum_{if}|\langle f|\mathbf{e}\cdot\hat{\mathbf{p}}|i\rangle|^2[f(E_i) - f(E_f)]\delta(E_f - E_i - \hbar\omega)$$

[$f(E)$ is the occupation number and Ω the volume of the system] and imaginary part is related through Kramers-Kronig relations. The (relative) complex dielectric function then reads $\varepsilon_d(\omega) = 1 - i\sigma(\omega)/\omega$, the refractive index is obtained as its square root, and finally the transmittance and reflectance can be calculated employing Fresnel relations.

However, for nanostructures the approach of homogeneous environment is often not appropriate. Instead, scattering and absorption of light are described by cross-sections C_{scat} and C_{abs}, defined as the power scattered and absorbed by the nanostructure when illuminated with a unit intensity. In the sparse system nanoparticles with the planar density n_S, the transmittance T is proportional to $1 - n_S(C_{scat} + C_{abs})$ and the reflectance R is proportional to $n_S C_{scat}$. When the nanostructures are embedded in a homogeneous matrix (which is common), the relations above hold for a relative transmittance/reflectance with respect to an empty matrix.

Luminescence spectroscopy is a technique in which the luminescent light is observed. Luminescence is the light produced by the sample besides the thermal equilibrium (black-body) radiation. There are several mechanisms for the excitation of the sample out of the thermal equilibrium: external light (photoluminescence), injection of the carriers from conductive leads (electroluminescence),

transfer of energy from high-energy beam of electrons passing through or near the excited structure (cathodoluminescence), etc. The photon energy of the luminescence is characteristic for the studied sample and independent of the parameters of excitation (e.g. the wave length of the exciting light).

The principle of the luminescence will be explained for the case of a semiconductor. An electron is excited from the valence to the conduction band, leaving a hole in the valence band. The system is now out of the thermodynamic equilibrium and will tend to restore it. The relaxation processes can be characterized as non-radiative (without the emission of photon) and radiative. Non-radiative processes (mostly phonon emission but also Auger processes, impurity scattering, etc.) are very efficient (time scale of ps or even fs) but allow only for a limited decrease of the electron's energy (tens of meV corresponding to a typical phonon energy). Radiative processes are generally much slower (time scale of ns) but allow for an arbitrary energy decrease (a photon of any energy can be emitted). Thus, the electron will first relax non-radiatively to the bottom of the conduction band (and the hole to the top of the valence band). The band edge presents so-called metastable state for which the non-radiative relaxation processes are no longer effective. The charge carriers dwell there for a comparably long time and eventually recombine through a radiative process, emitting a photon that contributes to the luminescence.

Luminescence spectroscopy is extremely sensitive technique, allowing in principle to register the emission of individual photons. It is however best suited for probing low-energy excitations, for which the non-radiative decay is hindered. It is also problematic to determine the strength of the transitions quantitatively because of complex relaxation effects influencing the recorded luminescence intensity.

1.4 Semiconductor quantum dots

Quantum dots (QDs) are semiconductor nanometer-sized heterostructures in which the electron motion is quantized in all direction and in consequence they exhibit discrete energy levels and wave functions confined to a finite volume. For that, QDs are also referred as artificial atoms. Quantum dots combine benefits of individual atoms (sharp density of states and slow decoherence) and solid state (easy integrability into more complex devices) and are therefore promising candidates for many applications such as lasers with low threshold current, single electron transistors, or single photon sources.

1.4.1 Fabrication

The methods for the fabrication of quantum dots can be divided into two classes – epitaxy and chemical synthesis.

Epitaxial quantum dots are prepared by depositing thin layers on a monocrystalline substrate. Two most common methods for the deposition are molecular beam epitaxy and metal-organic vapour phase epitaxy. [24] Typical growth is based on a strain-driven self-assembly and consists of several steps. On a substrate composed of a barrier material, a buffer layer of the same material is deposited to improve the quality of the surface. A thin layer (a few monolayers) of the QD material follows. Essentially, the two materials has to differ in the lattice parameter. The strain induced by the lattice mismatch is relieved by the formation of islands (future QDs). Depending on the growth details a thin layer of QD material can be left below the islands (Stranski Krastanov growth mode) or the islands are located right at the substrate (Volmer-Weber growth mode). To facilitate the island formation, the growth can be interrupted for a short time (tens of seconds). Finally, the structure is terminated by the upper barrier layer.

The growth of lattice-matched quantum dots, e.g. GaAs dots with AlGaAs barrier, requires modified approaches. Droplet epitaxy is a modification of molecular beam epitaxy, in which metallic droplets (Al) are formed on the bottom barrier. The sample is then annealed and the droplets etch the

underlying barrier. Nanoholes formed during this step are subsequently filled with the dot material (GaAs) and overgrown by the upper barrier. In this way, strain-free quantum dots are fabricated [26].

Chemically synthesized quantum dots are also known as colloidal quantum dots (for they are kept in solutions) or core-shell structures (for their characteristic shape of a core spheroid surrounded by spherical shell of the barrier material). They are mentioned here for completeness as the thesis deals solely with the epitaxial quantum dots.

1.4.2 Quantum confinement

QDs are composed of semiconductor crystals and electrons in them interact with a tremendous number of particles – lattice or other electrons. Therefore, we need a simplified view to describe quantum confinement in such a complex system in a comprehensible manner. The simplification is offered by the effective mass paradigm. Looking at the band structure of a bulk crystal we realize that the dispersion relation close to the band edges is parabolic, $E(k) = Ak^2$, and thus resembles the dispersion relation of free particles, $E(k) = (\hbar^2/2m_e) \cdot k^2$. The different slope can be treated as if the particles in crystal acquire effective mass m^* different from the mass of free electron m_e. The wave functions (Bloch waves) are composed of a plane wave part corresponding to the free particle and a periodic part. Taking these two facts into account it is natural to treat the crystal as a homogeneous environment of constant potential energy, in which electrons propagate as free particles. The effect of the complex crystal potential is hidden into modified effective mass and the modulation of the wave function by the periodic part of the Bloch wave. Such model constitutes the foundation for a description of heterostructures. Here, the band edge energies vary with the position and present a position-dependent effective confinement potential for the movement of the electrons with the (band dependent) effective mass. The confined wave functions have to be again multiplied by the periodic parts of the Bloch waves.

As an example, we can consider a quantum dot composed of GaAs (narrow band gap semiconductor, $E_g^{\mathrm{GaAs}} = 1.52$ eV at the temperature of 0 K) embedded in $Al_{40}Ga_{60}As$ (wide band gap semiconductor, $E_g^{\mathrm{AlGaAs}} = 2.06$ eV) [27]. The sum of the depths of potential wells for electrons (V_e) and holes (V_h) equals to the difference of band gaps, $V_e + V_h = E_g^{\mathrm{AlGaAs}} - E_g^{\mathrm{GaAs}} = 0.54$ eV. The determination of V_e and V_h is a tricky task and the data are usually not very reliable. Ref. [27] proposes $V_e = 0.34$ eV and $V_h = 0.21$ eV. The effective mass of the conduction band electrons in GaAs reads $m^* = 0.067 \cdot m_e$, the effective mass of the holes is anisotropic and cannot be described by a single value.

Two types of quantum dots are distinguished based on the band edge alignment of the inner (QD) and outer (barrier) material. For type-I QDs, both electrons and holes are confined inside the QD (V_e and $V_h > 0$). For type-II QDs, only one type of charge carriers is confined in QDs, while the other resides in the barrier and is only loosely bound to the QD by the electrostatic interaction with the former charge carrier, or by the inhomogeneous strain or piezoelectric field around the QD. Among QDs studied within the thesis, those based on GaAs/AlGaAs or InAs/GaAs materials belong to type I, Ge/SiGe are of type II, and InAs QDs with GaAsSb capping are of transitional character with the type of confinement dependent of the thickness or composition of the GaAsSb layer.

Quantum confinement results into discrete energy spectrum and localized states. The former feature is related to narrow (in theory delta-function) density of states and thermal stability (low-energy thermal excitations are suppressed due to separation of energy levels). The latter feature results into rather strong Coulomb coupling between individual single-particle states and stable excitons and excitonic complexes.

1.4.3 Characterization

Quantum dots are typically embedded in the barrier region, which makes their structural characterization difficult. Further, the strain usually imposed by the capping layer causes pronounced morphological changes during the overgrowth which excludes the possibility to determine the shape and dimensions of quantum dots prior their embedding. Still, AFM provides a useful insight into the morphology of quantum dots. It was used for example to study the overgrowth of a QD with the capping layer [28], showing a clear flattening and lateral elongation of the studied QD. Direct determination of the QD size and shape is possible for the strain-free GaAs QDs in the AlGaAs barrier, for which the shape is not expected to change after the overgrowth [26, 29]. X-ray diffraction presents a powerful tool, but rather difficult to use [30, 31].

Photoluminescence is the primary method to study the electronic structure of quantum dots, in particular their low energy excitations. As a non-destructive and non-contact method it allows for plenty flavors, including the possibility to apply external electric, magnetic, or strain field, study thermal and pumping-intensity dependences, or perform time-resolved studies. The spectral resolution of the technique can be below several tens μeV (i.e., 10^{-5} on the relative scale). With the spatial resolution about 1 μm it is possible to study individual QDs. Photoluminescence together with the interpretation based on the theoretical modeling provides confinement energies of electrons and holes, binding energies of excitonic complexes, electric dipole moments, tunneling energies in quantum dot molecules, radiative lifetime of excitons, indications for the lateral symmetry breaking, and many others. Some of these cases are discussed further in this thesis. Less frequently, electronic structure of quantum dots is studied by optical absorbance measurements. Scanning tunneling spectroscopy allows to visualize the wave function of the bound states together with the determination of their energies [32, 33].

1.4.4 Theoretical methods

As the theoretical modeling of quantum dots constitutes substantial part of the thesis, I briefly review the involved methods. Typical procedure consists of four steps: calculation of fields, single particle states, multiparticle states, and optical properties. Except for the highly symmetric instances or simplified models, numerical solutions are necessary.

The first step consists in the calculation of fields that affect the band structure of solid: strain, electric, and magnetic field. Strain field is present either as built-in field due to lattice mismatch or as external field. It is obtained by minimizing strain energy within continuum elasticity model, or, less frequently, within atomistic model, e.g. valence force field. Electric field can be present due to piezoelectricity, pyroelectricity, or as external field. It is calculated from Laplace equation. I note that the electric field produced by occupied confined states is usually not considered in single-particle calculations.

Single particle states are most often obtained using $\mathbf{k} \cdot \mathbf{p}$ theory. With some simplification, this method represents a multiband extension of the effective mass theory. The wave function is expressed as $\psi = \sum_b \chi_b u_b$, where the summation goes over a subset of energy bands, u_b is a periodic part of the Bloch wave at a specific point of Brillouin zone (usually Γ), and χ_b is a corresponding envelope function. Envelope functions are obtained from a set of Schrödinger-like differential equations and follow the normalization condition $\sum_b \langle \chi_b | \chi_b \rangle = 1$. Most frequently, eight bands are considered, two conduction and six valence (heavy hole band, light hole band, and spin-orbit split-off band, each doubled by a spin). The Hamiltonian is composed of Luttinger–Kohn Hamiltonian and Pikus–Bir Hamiltonian accounting for the effects of strain.

Alternatively, tight-binding or pseudopotential methods are used to obtain single-particle states.

In many quantum dots, including all instances considered here, Coulomb interaction between

individual particles represents only a small correction to the confinement. It is thus appropriate to obtain multiparticle states (excitons and multiexcitonic complexes) by expanding the wave function into a series of limited set of Slater determinants formed from single particle states. This approach is named configuration interaction.

More details and references to this part is provided e.g. in [34, 35] and references therein.

1.5 Plasmonic antennas

Electrons, represented by plane waves in free space or by Bloch waves (modulated plane waves) in a bulk crystal, can be confined to a finite volume by the heterostructures with offsets in their band edges. Similarly, photons (light) represented by plane waves in free space can be confined to a finite volume by heterostructures with a contrast of their refractive indices. Such a heterostructure can be realized in small metal particles embedded in a dielectric environment, such as insulators, semiconductors, or air.

The mechanism of the light confinement in small metal particles is closely related to the quasiparticles called localised surface plasmons. Bulk metal supports collective oscillations of the free electron gas. Bulk plasmon is a quantum of such oscillations. On the boundary of the metal and a dielectric, the oscillating electrons produce a macroscopic electric field, evanescent waves that are free to move along the boundary and decay exponentially away from the boundary. The quasiparticles arising from the strong coupling between some excitation and the electromagnetic field are named polaritons. Here, so-called surface plasmon polaritons (SPP) are formed, referring to the coupling of the plasmon oscillations on a surface of metal to the evanescent wave propagating along the surface. Surface plasmon waves can be described by their frequency and by the in-plane component of the wave vector. The dispersion relation has two branches, whose asymptotes are the light dispersion line in the surrounding medium and two constant frequencies corresponding to the bulk plasmon and surface plasmon frequency [36]. The coupling is manifested by the avoided crossing of the lines. The electromagnetic field component of SPP is confined in the direction perpendicular to the metal surface but no in-plane confinement is observed.

When the metal surface is spatially restricted (such as in the case of metal nanoparticles), the interference of propagating surface plasmon waves gives rise to the standing waves; the related quasiparticle is called localised surface plasmon (LSP). The dispersion relation is now composed of discrete points, corresponding to the localized surface plasmon resonances (LSPR). Due to high losses in metal, the lifetime of the resonances is very short (femtoseconds) and their spectral width is thus rather large. Consequently, the response of the system supporting localised surface plasmons is spectrally continuous, with distinct peaks corresponding to different LSPR only if the frequency separation of the modes is larger than their spectral width (typically the case of several low-frequency LSPR in supported by small metal particles). The field related to LSPR is confined also in the in-plane directions.

For the experimental characterization of LSPR a variety of methods is employed. We describe here two which are standardly used at the Institute of Physical Engineering BUT. Optical spectroscopy is one of the simplest and commonly used techniques. In the transmission mode it provides the extinction cross section of plasmonic particles and in the reflection dark-field field mode it allows to study the scattering cross section (a special dark-field objective is used to collect the scattered light while blocking the directly reflected light). Due to its far-field nature, the spatial resolution of the optical spectroscopy is set by the Abbe diffraction limit and is thus insufficient for the imaging of local LSPR properties (such as the plasmon's near field distribution). Further, the method is blind to so-called dark plasmons. The electric field of dark plasmons has a vanishing dipole moment which impedes their coupling to the far-field radiation. The electron energy loss spectroscopy allows to

overcome both limits of the optical spectroscopy. In this technique, the sample in the form of a thin membrane (tens of nanometer) is scanned by a monochromated and focused beam of high-energy electrons (above 60 keV). The probe electrons interact with the electron gas in metal particles and can with some (very small) probability excite a LSPR. This is accompanied by a transfer of energy from the electron to a plasmon. If one detects the energy distribution of transmitted electrons, characteristic peaks at the energies corresponding to LSPR are observed. The spatial resolution of the method is better than 1 nm which allows for local imaging of LSPR. Further, the electrons interact also with dark plasmons. The spectral resolution of the method is about 0.01–0.1 eV – a fair value considering the energy of impinging electrons but naturally worse than the resolution of the optical spectroscopy. Further, the observation of the low-energy features (below 0.1–1 eV) is hindered by the strong zero-loss peak which corresponds to the electrons that pass through the sample without any inelastic scattering (it has a finite width due to a finite resolution of the monochromator).

The applications of localised surface plasmons are usually connected with their ability to focus and/or enhance the electromagnetic field. There are various plasmon-enhanced spectroscopic techniques, in which the studied sample is decorated with plasmon-supporting nanoparticles to enhance the measured signal. For example, plasmon-enhanced photoluminescence allowed to detect the fluorescence of individual molecules [37]. Another techniques comprise the tip or surface enhanced Raman scattering, surface enhanced infrared absorption, and others.

2 Quantum dot properties

This section is dedicated to general properties of quantum dots while more specific topics are discussed in the following sections. The research was triggered by design of complex quantum dot structures, development of a new tuning technique, or expanding the expertise gathered for standard InAs QDs to more exotic QDs. The studies have often introductory character, report rather basic properties of particular quantum dots, and their results are repeatedly used in the follow-up studies which pursue particular tasks of contemporary physics of semiconductor quantum dots.

2.1 Quantum dots with tunable thickness of the wetting layer

Strain-free GaAs quantum dots embedded in an AlGaAs barrier represent an unusual quantum dot system. The lattice parameter mismatch between both constituents is about 0.1 %, considerably smaller than e.g. in traditional InAs QDs embedded in GaAs (7 %). For simplicity and also for the sake of scientific marketing, the related low internal strain is advertised as zero internal strain, to which we will also adhere in the forthcoming discussion. In consequence, these QD are versatile as there is no strain-related restriction to their volume or shape. Further, the shape and composition of the QDs can be experimentally retrieved with the accuracy and reliability impossible in any strained QDs: (1) There is negligible intermixing between the QD and barrier material during the growth. (2) Change of the QD shape during the growth of the capping layer (i.e., top barrier), in particular decrease of the height, which is common in strained QDs [28, 38], is absent here – this allows for easy determination of the QD morphology on uncapped samples. Together with simple form of the quantum confinement, which is not contributed by the strain or piezoelectric field, this facilitates the interpretation of the experimental data with reliable theoretical models. The GaAs/AlGaAs QDs are thus ideal testing system for new physical ideas and concepts. In particular, they are suitable for the studies of the excitonic fine structure splitting and its reduction towards zero (Sec. 3.1) as the total anisotropy of the QD is dominated by the structural anisotropy and the piezoelectric contribution is not present.

On the other hand, zero internal strain hinders the fabrication of the GaAs/AlGaAs QDs as the strain-driven self-assembly cannot be employed. Instead, complex growth procedures were developed including local droplet etching [39], modified droplet epitaxy [40], or hierarchical self-assembly [41].

In our study [29] the hierarchical self-assembly [41] was used for the GaAs/AlGaAs QD fabrication. In first step, InAs QDs were prepared by molecular beam epitaxy using the usual Stranski-Krastanov growth mode and capped by 10-nm thick GaAs layer. Next, strain-sensitive etching by $AsBr_3$ gas resulted in removal of InAs QDs and spontaneous formation of nanoholes in the GaAs layer at the place of InAs QDs. The bottom barrier layer of $Al_{0.45}Ga_{0.55}As$ was deposited at low temperature so that the nanohole was transferred to AlGaAs surface. A thin layer of GaAs was deposited and the sample was annealed so that GaAs filled the nanohole. The excessive amount of GaAs, if present, formed a thin layer (an analogue of a wetting layer, WL) above the filled nanohole (quantum dot). The structure was completed with the top $Al_{0.35}Ga_{0.65}As$ barrier. Due to anisotropic surface diffusion of Ga and Al atoms the resulting dots were strongly elongated along [110] direction, a fact which is important for the studies of fine structure splitting described in Sec. 3.1.

A series of samples with a different thickness of the wetting layer d (between 0 and 4 nm) was prepared and characterized by low-temperature photoluminescence with microscopic spatial resolution. Emission from quantum dots and a wetting layer was identified in the spectra, both exhibiting a pronounced red shift with increasing d (which corresponds to an increase of both the QD height and the WL thickness). With increasing excitation power, a shell-filling and the emission involving excited electrons and holes were observed.

The QD ground state emission was composed of several lines, a strong one at the high-energy side of the spectra and several weaker red shifted by $\geq$ 2 meV. With the help of polarization-resolved measurements and excitation-power-dependent measurements, the former was identified as neutral exciton (X) recombination, while the latter lines arose from the biexciton (XX, polarization doublet), positive trion (X^+, polarization singlet), or negative trion (X^-, polarization singlet). The discrimination between both trions was based on the background p-doping resulting in higher intensity of X^+ in comparison to X^-. The bonding nature of both trions and biexciton is rather uncommon in strained QDs and suggests a large overlap of electron and hole wave functions. This is facilitated by the absence of the piezoelectric field (which tends to separate the carriers with opposite charge) but still surprising due to a large effective mass difference between electrons and holes. We have used the concept of effective confinement potential [42] which is steeper for lighter electrons and shallower for heavier holes to show that in total the extension of both wave functions is comparable. Further, the biexciton and trion binding energies are only weakly dependent on d. As d the increases, the wave functions become more extended and the Coulomb interaction weakens (which tends to decrease the binding energies), but at the same time the correlation becomes more pronounced (which tends to increase the binding energies) and both the effects almost cancel each other.

The measurements are well reproduced and interpreted with the numerical simulations combining the 8-band $k \cdot p$ theory for single particle states with the configuration interaction method for the excitonic complexes. In conclusion, a simple way to modify the height of the strain-free QDs and tailor their properties was demonstrated together with some peculiar properties such as the ultimate overlap of the electron and hole wave function and related bonding nature of all exciton complexes. These QDs were used in subsequent studies of the excitonic fine structure splitting [43, 44, 45] (see Sec. 3.1).

2.2 External strain as a tool for tuning QD properties

The strain field is an important ingredient in determining QD properties. Inhomogeneous internal strain is present in lattice-mismatched QDs with a magnitude up to several percent.External strain (usually homogeneous on a scale of QD dimensions) can be imposed to engineer the QD properties; the magnitude of such strain is considerably weaker than in the case of the internal strain and reaches usually up to 0.1 %. Unintentional external strain can result for example from the presence of defects in the barrier or vicinity of the cleaved edge of the sample.

The effect of the strain on the band structure is described by the Pikus-Bir Hamiltonian [46] and comprises the band shift and mixing. Elastic potentials are introduced to describe the magnitude of the shift. They refer to the shift per unit component of the strain tensor (or their linear combination) and their characteristic values range between 1 and 10 eV. For example, the heavy and light hole band in InAs QDs embedded in GaAs are split by about 0.4 eV and the fundamental band gap increases from 0.4 eV to more than 0.6 eV due to strong internal strain. The shifts induced by the external strain field are considerably smaller (up to several tens of meV) but still sufficiently large to induce observable effects e.g. in modifying the biexciton binding energy [47]. Band mixing is important in for the exciton polarization properties [45], exciton fine structure splitting [44], or making the dark exciton states optically active [48].

We have employed the external strain field in several studies aiming at the tuning of various quantum dot properties including the biexciton binding energy [47] (Sec. 2.2), the magnitude of fine structure splitting, polarization of the exciton transitions and the polarization anisotropy of QD photoluminescence [44, 45] (Sec. 3.1) or the tunneling energy in QD vertical molecules [49] (Sec. 4.2).

The principle of the strain application is common to all the studies, although details may differ due to technological progress over time. In the following we describe our state-of-the-art setup for strain application [50, 51]. The sample is prepared in a form of a nanomembrane containing the QDs, possibly embedded in a p-i-n diode structure if the simultaneous application of the external electric field is desired. The nanomembrane is integrated onto a lead magnesium niobate-lead titanate $[Pb(Mg_{1/3}Nb_{2/3})O_3]_{0.72} - [PbTiO_3]_{0.28}$ (PMN-PT) piezoelectric actuator by gold-thermocompression bonding. PMMA resist used to bind the nanomembrane to the piezo-actuator in earlier works allowed to transfer only a limited strain and was prone to damage. The voltage applied on the piezo-actuator induces the in-plane biaxial strain field, either tensile or compressive depending on the sign of the electric field. The strain is transferred to the QD sample depending on its position on the actuator either as in-plane biaxial (the QD membrane on top of the actuator) or as uniaxial with the principal strain axis usually aligned with the QD [110] direction (the QD membrane at the side of the actuator). In the latter case, denoting the strain along the principal axis ε, the strain along the perpendicular axis reads $-0.7 \times \varepsilon$ [52]. The method allows to impose the external strain up to 0.4 % [50].

In one of our early studies employing the external strain field we have addressed the binding energies of exciton species in InAs/GaAs QDs. Thin GaAs membranes (200 nm) with a single layer of self-assembled InGaAs QDs were fabricated using molecular beam epitaxy, transferred onto piezo-actuators and characterized by low-temperature photoluminescence with the spatial resolution allowing to focus to individual QDs. Typical spectra consist of three sharp lines attributed to the recombination of the exciton (X), positive trion (X^+), and biexciton (XX). The assignment of the lines was based on the pumping-power and polarization dependent measurements. The transition energy of the neutral exciton was about 1.37 eV which gives evidence of rather small QDs. In different QDs, both red shift and blue shift of the biexciton with respect to the exciton was observed while the positive trion was consistently blue-shifted.

Binding energy E_B of the excitonic complex Y is the difference between the energy of complex

components (a neutral exciton and a hole for the positive trion and two excitons for the biexciton) and the complex itself. The transition energy E_{tr} of that complex (i.e., the energy observed in the photoluminescence spectra) is the difference between the energy of the complex and the energy of the final state after the recombination of single electron-hole pair (i.e., an exciton for the biexciton and a single hole for the positive trion). The transition energy of the complex Y can be calculated by subtracting its binding energy from the transition energy of the neutral exciton, i.e.,

$$E_{tr}(Y) = E_{tr}(X) - E_B(Y).$$

The complexes with the positive/negative binding energy are called binding/antibinding and their spectral line is red-shifted/blue-shifted with respect to the neutral exciton line. In the studied QDs, the binding energy of the biexciton ranged from −2 meV to 2 meV and the binding energies of X^+ ranged from −8 meV to −3 meV. We note that the binding energy is not a measure of the complex stability – the components are held together by the quantum confinement, not by Coulomb interaction. In this aspect the exciton complexes in quantum dots differ considerably from those in a bulk crystal or quantum wells and wires.

The energies of exciton complexes can be estimated from the single particle energies (E_e for the ground electron state and E_h for the ground hole state) and the Coulomb integrals between those ground states (a negative J_{eh} describing the electron-hole attraction and positive J_{ee} and J_{hh} for the electron-electron and hole-hole repulsion, respectively). Such simple approach provides an elementary insight into the exciton complex properties, although it fully neglects the self-consistency (a modification of the wave functions due to the additional Coulomb potential) and correlation (a collective behavior of individual particles forming the complex) effects. For example, biexciton is composed of two electrons and two holes with four e-h attractions and one e-e and one h-h repulsions. Its energy reads

$$E(XX) = 2E_e + 2E_h + 4J_{eh} + J_{ee} + J_{hh}.$$

The energy of exciton reads

$$E(X) = E_e + E_h + J_{eh}$$

and for the binding energy of the biexciton we thus obtain

$$E_B(XX) = 2E(X) - E(XX) = -2J_{eh} - J_{ee} - J_{hh}.$$

Similarly, the binding energy of the positive trion reads

$$E_B(X^+) = -J_{eh} - J_{hh}.$$

Strongly antibinding positive trion therefore requires that the hole-hole repulsion is considerably stronger than the electron-hole attraction, $|J_{eh}| < J_{hh}$. This is possible when the hole wave function is more compact than the electron wave function or when both particles are spatially separated (i.e., having different average coordinates). The latter case would result into strongly antibinding biexciton which is not observed. From the binding energies we therefore conclude that the hole is confined into a smaller volume than the electron (which is consistent with its larger effective mass).

When the external biaxial strain was applied to the QDs, the exciton transition energy $E_{tr}(X)$ was shifted up to several meV (blue-shifted for the in-plane compression and red-shifted for the in-plane expansion). Interestingly, the binding energies of XX and X^+ show a linear increase with $E_{tr}(X)$ with a slope

$$\gamma(Y) = \Delta E_B(Y)/\Delta E_{tr}(X)$$

(Y stays for XX or X^+), which for different QDs acquires values between 30 μeV/meV and 70 μeV/meV, similar for both species.

The observations were explained with the help of theoretical modeling using the empirical pseudopotential calculations or the $k \cdot p$ calculations for single particles states combined with the configuration interaction calculations of the exciton states. The increase of the transition energies upon in-plane compression was explained by the shift of single particle energies. The observed changes in the binding energies were explained by the modification of the Coulomb integrals. The electron wave function tends to be more localized upon compression, which resulted in enhanced magnitudes of J_{ee} and J_{eh}. At the same time, the hole wave function becomes slightly less localized but the effect on J_{hh} was considerably weaker. Considering the weight J_{ee} and J_{eh} in the binding energy formulas, this explains the increase of the binding energies $E_B(XX)$, $E_B(X^+)$ upon compression.

For a QD with originally low biexciton binding energy it was demonstrated the the emission energies of X and XX can be tuned by the external strain to the same value. This has an interesting application in the production of polarization-entangled photon pairs utilizing so called time-reordering scheme proposed by Avron *et al.* [53].

2.3 Quantum dots between type I and type II confinement

The QDs with the type-II confinement (one type of the charge carriers is localized in the QD volume while the other in the barrier material, usually close to the QD) have considerably reduced overlap between the electron and hole wave functions. Consequently, the spontaneous emission rate is low and the radiative life time of excitations is high compared to type-I QDs [54]. This is for instance exploited in QD flash memories [55, 56]. In solar cells, QDs are used to enhance the infrared spectral response; the type-II QDs allow for an easier charge extraction [57, 58]. Even in purely optical applications such as lasers the InAs QDs with the GaAsSb overlayer close to the transition between type-I and type-II confinement are studied as their emission fits into the communication wavelengths of quartz optical fibers (1.3 and 1.55 μm) [59, 60]. Among several dot/barrier material combinations (e.g., Ge/Si, GaSb/GaAs, CdTe/CdSe, etc.), InAs QDs capped with a ternary $GaAs_{1-y}Sb_y$ layer are particularly interesting because of the possibility to smoothly tune the QD confinement between type I and type II by setting a proper Sb content or the thickness of the GaAsSb layer. In this system, electrons are confined inside the QD and the holes reside in the GaAsSb layer.

Our interest in the GaAsSb capped QDs was originally triggered by the experimental effort to red-shift the emission wavelength of the standard InAs QDs towards the infrared telecommunication wavelengths of 1.3 and 1.55 μm. The role of the GaAsSb overlayer was to relieve the internal strain and to reduce the potential barrier. The reduced strain inside QDs results in a direct red shift of the emission and can allow for a growth of larger and taller QDs accompanied by additional redshift; the latter effect can be considerably larger than the former one [61]. In our theoretical studies [62, 63], we have successfully reproduced previous experiments and interpreted the red shift of the emission with increasing Sb rate in the overlayer as a joint effect of the reduced strain field and a shifted band edge resulting in the reduced height of the confinement barrier in type I and reduced fundamental band gap in type II. In type I, both effects of the reduced strain and shifted band edges were equally important, in type II the latter prevails. Interestingly, no increase of the QD volume was found in contrast with the QDs with InGaAs overlayer [61]. This observation was attributed to the surfacting effect of antimony atoms which helps to preserve the shape and the height of the QDs during the overgrowth.

Type-II QDs exhibit unusual properties different from type I QDs which give rise to non-trivial physical phenomena such as the non-linear Stark effect, formation of molecular states, or vanishing fine structure splitting (see Sec. 3.2) despite the low symmetry of QDs. These phenomena are connected with the position and shape of the hole wave function. The hole confinement potential profile inside the GaAsSb overlayer is determined by the inhomogeneous elastic strain, piezoelectric

potential, and, if present, composition and thickness fluctuations of the layer. The principal strain component contributing to the shift of the heavy-hole band edge is the biaxial strain B expressed in terms of the strain tensor components ε_{ij} as

$$B = \varepsilon_{zz} - (\varepsilon_{xx} + \varepsilon_{yy})/2$$

(z is perpendicular to the QD plane in accordance with the standard notation). The shift reads $\Delta E_{hh} = -b \cdot B$ where b is the biaxial deformation potential. Since b is negative, the holes are preferentially located in the regions with large positive B, i.e, in regions with the in-plane compression and the vertical expansion, which are located at the sides of a QD. A QD material has a largest lattice parameter of all the structure and acts therefore as a stressor. The overlayer at the sides of the QDs tends to be compressed vertically and stretched laterally. The vertical position of region of large positive B is rather sensitive to the In distribution inside the QD.

The piezoelectric field has an octopole form with the nodal plane slightly above the QD basis. Its magnitude is several tens meV, comparable with the strain-induced variations of the confinement potential. Exact position and shape of the hole wave functions therefore depends on the delicate interplay between both effects. In most cases, the piezoelectric field splits the hole wave function into two segments. The segments are either located at the dot basis and situated along [110] or located above the nodal plane of the piezoelectric field and situated along $[1\bar{1}0]$. The consequences of this unusual and intriguing hole wave function shape are discussed in the following.

Molecular states The confinement potential energy for electrons and holes is contributed by the piezoelectric potential energy, which exhibits four minima for the confinement of holes (along $[11\bar{1}]$, $[1\bar{1}1]$, $[\bar{1}11]$, and $[\bar{1}\bar{1}\bar{1}]$ direction from the center of a QD) and four minima for the confinement of electrons along the complementary directions. For electrons, the piezoelectric contribution means rather a small modification of the band-offset contribution and only contributes to a weak elongation of the electron wave functions. For the holes, the piezoelectric contribution is strong enough to split the wave function into two segments. Two lowest QD hole states then resemble bonding and antibonding molecular orbitals, although they can be in principle described as s and p atomic orbitals. The barrier between the minima of the potential energy is sufficient to produce true molecular states, with negligible probability density in the region between the segments. The width of the barrier can be adjusted at will by changing the lateral dimension of the QD. The (potential) height of the barrier is not tunable so easily but can be increased in some extent by increasing the height of the QD, which results into stronger strain field and thus stronger piezoelectric potential. Clear disadvantage of the concept resides in the impossibility to form the electron molecular states. Still, hole molecular states can be utilized in the Burkard's proposal of quantum gate device [64].

Non-linear Stark shift The hole wave function is usually located at the bottom two minima of the confinement potential, i.e., below the electrons. Thus, the electric dipole moment is negative (pointing downwards). When the positive electric field is applied (with a positive potential at the bottom of the quantum dot), a linear Stark shift (an increase of the exciton transition energy) is observed with a weak quadratic part (due to a small polarizability of rather strongly confined charge carriers). For a certain critical value of the field (100 kV/cm for the case studied in Ref. [62]) the confinement energy minimum above the QD becomes more favorable and the hole is located there, above the electron. This is accompanied by an abrupt change in the electric dipole moment (which is now positive, i.e., pointing upwards) and the change in the slope of the energy-field relation. The Stark shift energy-field curve thus consists of two regions of ordinary, almost linear Stark shifts with an abrupt change of the slope at the boundary of the regions.

Tunable fine structure splitting Fine structure splitting in type-II QDs is in more detail covered in Sec. 3.2.

3 Excitonic fine structure

Single particle states in quantum dots are spin-degenerate doublets, unless a magnetic field is applied. This fact follows from Kramers theorem [65] which states that in any system with the half-integer total spin and underlying to the time reversal symmetry, the eigenstates are at least doubly degenerate. Time reversal symmetry is a symmetry of physical laws under the time reversal transformation $t \rightarrow -t$. Schrödinger equation fulfils the time reversal symmetry unless the magnetic field is applied. Thus, the single particle states in quantum dots form degenerate Kramers pairs (the states differing only in a spin projection). Similarly, trions (charged excitons, particles composed of two electron and one hole or one electron and two holes) form degenerate Kramers pairs. On the other hand, excitons are integer spin particles and as such they do not follow Kramers theorem. In Hartree approximation, where only a direct Coulomb interaction is considered, excitons are degenerate quadruplets. The exchange interaction between the electron and the hole splits the exciton levels, giving rise to a so-called fine structure. In common QDs, two of those levels are *bright* (with a short radiative lifetime and a strong optical response, e.g., photoluminescence) and two are *dark* (with a long radiative lifetime and a weak optical response, often below the experimental detection limit). The existence of such levels can be explained by a simple model. The conduction band electrons have the secondary (projection) total angular momentum quantum number $m_J^{(e)} = \pm 1/2$ while for the heavy hole states the number reads $m_J^{(h)} = \pm 3/2$. For exciton, $m_J^{(X)} = m_J^{(e)} - m_J^{(h)}$ holds, i.e., $m_J^{(X)} = \pm 1,\ \pm 2$. As the spin number of photon is 1, the selection rule for the single photon processes reads $\Delta m_J = 0,\ \pm 1$. The excitons with $m_J^{(X)} = \pm 1$ can recombine into the vacuum state ($m_J = 0$) producing single circular-polarized photon – they are therefore bright. On the other hand, excitons with $m_J^{(X)} = \pm 2$ cannot recombine by any single photon event (because the photon with a spin of 1 cannot carry the exciton momentum) – such excitons are dark. These simple rules are only approximate in the realistic QDs. For example, the heavy-hole – light-hole (hh-lh) mixing changes m_J of the hole states and makes the dark states optically active, although not as intense as the bright states. Such mixing is naturally present in QDs due to quantum confinement and can be also induced and tuned by the external strain field.

Considering the energy, the exciton quadruplet is split into dark and bright pairs. The dark pair has usually lower energy and is almost degenerate. The bright pair, usually of higher energy, is further split; this splitting is known as fine structure splitting (FSS). Dark-bright splitting arises from the symmetric part of the exchange interaction and is thus a universal feature of quantum dots. FSS is attributed to the asymmetric part of the exchange interaction, it is thus present in QDs with reduced symmetry (lower than D_{2d} [66], an example is an elongated QD).

FSS was observed in various QD systems with the magnitude between zero [26] (which corresponds to values below a few μeV considering the experimental resolution) and 1 meV [67]. The theory of FSS was developed by Takagahara [68] soon after its discovery and further elaborated by numerous authors, our approach is briefly presented in Ref. [34]. The interest in FSS is both practical and fundamental. FSS helps to identify the optical transitions originating in the exciton, trion, and biexciton recombination. It plays an important role in the spin initialization e.g. in electron spin memories [69] or in exciton dynamics and dephasing. [70] It provides some insight into the size and shape of QDs. Benson's proposal of the source of entangled photon pairs relying on zero FSS [71] has called for the preparation of QD systems with low FSS. This issue was addressed in numerous studies either by sophisticated tailoring of the fabrication procedure to produce the desired high-symmetry QDs, or by applying a post-growth treatment or external field to tune the FSS.

Several our studies focused on FSS in various QD systemswith the aim to understand the fundamental properties of the bright exciton doublet and consider its utilization in the polarization-entangled photon pair source. GaAs QDs with AlGaAs barrier are addressed in Sec. 3.1, and type-II InAs QDs with a thin GaAsSb overlayer and a GaAs barrier in Sec. 3.2. Finally, a recent study on

dark excitons is briefly described in Sec. 3.3.

3.1 Fine structure splitting of AlGaAs quantum dots

In a series of papers [43, 44, 45] we have studied elongated strain-free GaAs QDs with AlGaAs barrier described in more detail in Sec. 2.1. In the last paper, the quest for zero FSS was pursued by adjusting the growth process so that the resulting QDs have a symmetric (circular) shape.

In the first paper [43], we provided a direct quantitative comparison of measured and calculated FSS. This was enabled by using strain free QDs whose dimensions, shape, and composition were experimentally determined with then unprecedented accuracy. FSS was determined from the polarization-resolved photoluminescence of individual QDs. The spatial resolution of the setup allowed to collect the luminescence from as few as several tens of QDs. Since individual QDs emitted narrow lines (recorded linewidth of about 70 μeV is determined by the spectral resolution of a microphotoluminescence setup) and the ensemble emission covered the spectral range of about 10 meV (due to QD shape and size inhomogeneity), it was possible to resolve the emission of individual QDs. FSS was determined as the extent of characteristic wavy patterns formed by the peak energy of the emission plotted as a function of the polarization angle. The values determined for different structures ranged from 10 to 200 μeV. Simulations for realistic QD model (using as input scanning probe microscopy data of QD structures) reproduced both magnitude of FSS and polarization of both components of the emission with a reasonable accuracy.

Due to the elongation of QDs, the values of FSS are too large for the source of polarization-entangled photon pairs. For this reason, in our second study we explored the possibility to tune FSS by the external strain field [44]. Thin MBE-grown membranes (about 150 nm) containing GaAs/AlGaAs QDs were released from the substrate (by selectively etching off a sacrificial AlAs layer) and transferred onto a side face of a piezoelectric actuator (PMN-PT crystal). The voltage applied to the actuator results into strongly anisotropic biaxial strain with components $\varepsilon_{\parallel}$ and $\varepsilon_{\perp} \approx -0.7\varepsilon_{\parallel}$ along the direction parallel and perpendicular to the electric field, respectively. The membranes were oriented so that the electric field direction was close to the $[1\bar{1}0]$ GaAs crystal direction, deviating by an angle up to 20°. When sweeping the strain $\varepsilon_{\parallel}$ in the range of few ‰(from negative to positive values), FSS first decreases and then increases. At the same time the polarization of the high-energy component of the excitonic emission changes by almost 90° (51–79° for different QDs), being almost perpendicular to the pulling direction for large strain. These observations are consistent with a strain-induced anticrossing of the bright exciton states.

The effect is reproduced and explained by a theoretical modelling. Strain field results into the mixing of heavy and light hole bands, yielding the bands with anisotropic effective mass. In turn, the elongation of the hole wave function significantly changes, becoming larger in the direction of the compressive strain. Strain can even compensate for or preponderate over the structural elongation, resulting into circular or inversely elongated hole wave functions in elongated QDs. At the same time, the electron wave function is only negligibly modified.

When a QD elongation axes coincide with the strain axes, a crossing of bright exciton states is predicted with a zero FSS and abrupt change in the polarization of the high-energy component's emission. For a tilt between both axes (and also in a realistic case of irregularly shaped QDs) there is an anticrossing with a finite minimum FSS and a smooth change in the polarization. The latter case corresponds to the experimental observations. Interestingly, in that case the hole wave function can be not only elongated, but also rotated.

In the third study the aspects of the strain-tunable FSS are discussed more thoroughly, including the relation between the heavy-hole–light-hole mixing, FSS, and polarization orientation and polarization degree of the excitonic emission [45].

3.2 Fine structure splitting of type-II quantum dots

Our theoretical study [34] dealt with the InAs QDs with a thin GaAsSb overlayer (GO) further embedded in GaAs. Depending on the thickness and the composition (Sb content) of the GO, the confinement can be continuously varied between type I and type II with the holes confined inside the GO in the vicinity of the QD. The transition between type I and type II is accompanied by a change in the hole wave function shape, in particular, in type II a two-segment hole wave function is formed. As we have learned in the foregoing, FSS is closely related to the elongation of the wave functions. These facts open an interesting prospect for tuning the FSS which is explored in the study. In particular, it is shown that small values of FSS comparable with the natural linewidth of the excitonic transitions are achievable for realistic quantum dots just by setting suitable GO properties.

We have studied the effect of the segmentation of the hole wave function with a toy model where the electron and hole probability densities were represented by anisotropic Gaussians. Splitting of the hole wave function into two segments resulted in a modification of its effective elongation with a possibility to compensate the electron elongation and achieve zero FSS for a range of parameters. The effective elongation is contributed by both the elongation of the segments and their mutual position.

Our inspection of the effective confinement potential identified the inhomogeneous strain field and the piezoelectric field as key ingredients determining the shape of the hole wave function. In particular, the piezoelectric field has a form of an octopole with a nodal plane slightly above the basis of a QD. Consequently, changing the vertical position of the hole shall result in a pronounced change in the position and shape of the segments.

In the next step we have attempted to vary the thickness of the GO to see whether we can vertically shift the hole wave function and modify the FSS. The electron wave function resided inside the QD and was almost unaffected. We indeed observed the expected effect (more complex than a simplified explanation presented here) and for three considered model QDs (a lens, an elongated lens and a pyramid), zero FSS was achieved for a certain thickness of the GO.

Further, we have calculated the exciton radiative lifetime to see whether these type-II QDs are suitable for optical applications, in particular for the source of polarization-entangled photon pairs. In type-I QDs, typical lifetimes are about 0.5 ns. The zero FSS was obtained for the structures with the lifetimes of 0.9 ns and 2.2 ns (lens), 3.4 ns (elongated lens), and 4.8 ns (pyramid). Thus, these type-II QDs are still rather good emitters. We concluded that low natural FSS and efficient photoluminescence make the GaAsSb capped InAs QDs attractive as a possible source of entangled photon pairs.

In the follow-up study [72] we have shown that similar results can be obtained when the transition between type-I and type-II confinement is induced by the variation of the Sb content in the GaAsSb overlayer of constant thickness.

The experimental verification of our predictions was not possible so far and will be attempted in the ongoing research.

3.3 Spontaneous brightening of dark exciton

Although the dark pair of the exciton quadruplet attracts less attention than its bright sibling, it is still of considerable interest. Most importantly, it exhibits long radiative lifetime which makes it attractive for spin storage for quantum information processing [73]. Further, the knowledge of dark-bright energy splitting provides an insight into the nature of the exchange interaction (including e.g. screening) and helps to retrieve the QD size and composition.

Dark excitons are known to emit light due to the valence band mixing effects [74]. The factors promoting the mixing and making the dark excitons optically active include external magnetic field,

external or built-in strain field, or shape asymmetries. The light propagates in the plane of a QD and is polarized along its vertical axis, different from in-plane polarized emission of bright excitons.

In our study [48] we reported the observation of dark exciton emission in strain-free GaAs/AlGaAs QDs with highly symmetric shape (identical to the last case of Sec. 3.1). The side emission from a cleaved facet of the sample was investigated by polarization-resolved micro-photoluminescence. Light was collected along $[1\bar{1}0]$ direction. Typical spectra consist of a strong [110]-polarized peak corresponding to one bright exciton and a weaker [001]-polarized peak corresponding to a dark exciton. The other bright exciton was observed indirectly due to total internal reflection within the sample and the second dark exciton remained dark.

In highly-symmetric QDs under no external field the brightening of dark exciton is not anticipated. The only symmetry-breaking factor is the presence of the cleaved facet. This may introduce a local electric field or an anisotropic strain field with major axis parallel to $[1\bar{1}0]$ direction. Our simulations have shown that it is this strain field (related e.g. to the oxidation of AlAs layer present below QDs) that is responsible for the brightening of the dark exciton. We also predict an increase of FSS (i.e., energy splitting of the bright exciton pair) in the vicinity of the cleaved edge due to the related strain, which was verified by subsequent measurements.

It is instructive to explain the mechanism of the brightening. As we have already mentioned, it is connected with the valence band mixing effect. The ground hole state is always dominated by the heavy hole band but it contains also a small contribution of light holes. Interestingly, there is a fair amount of light holes even in as-grown QDs. For a spin-up ground hole state, the weights of the light-hole-spin-up and light-hole-spin-down bands read 4.1 % and 0.7 %, respectively. However, in optical spectra there is no signature of light holes (such as the out-of-plane-polarized emission of the dark exciton). When the strain is applied (the magnitude of 0.1 % along the principal $[1\bar{1}0]$) axis has been set), the weight of the light holes increases moderately to 5.0 % and 2.0 % for spin-up and spin-down band, respectively. Now, the dark exciton is brightened. The reason for such a behavior consists in the symmetry of the mixing terms and the related symmetry of the envelope functions. The mixing terms between the heavy-hole and light-hole bands in the kinetic Luttinger-Kohn Hamiltonian [75] are differential operators that results into light-hole envelope functions quasi-antisymmetric in two coordinates, with negligible contribution to the transition matrix elements. On the other hand, the mixing terms in the strain Pikus-Bir Hamiltonian [46] are numbers that result into quasi-symmetric light-hole envelope functions with substantial contribution to the transition matrix elements. More details are provided in our study [48].

Let us now discuss the practical implications of our study. We have shown that even a cleaved edge suffices to make a dark exciton optically active. On one hand, this allows a simple access to dark excitons. On the other hand, the effect has to be taken into account when a long lifetime of dark excitons is desirable. We also propose the possibility to have both optically active and inactive dark excitons on a single sample. Finally, we envision the possibility of dynamical brightening/darkening of a dark exciton via application of anisotropic stress pulses.

4 Quantum dot molecules

Similar to atoms, two or more quantum dots (artificial atoms) can be positioned close to each other so that their electron orbitals overlap forming joint molecular states. Unlike ordinary molecules, quantum dot molecules (QDMs) are held together by the embedding medium rather than by bonding electrons. This allows to tailor the strength of the interaction as well as to form unusual molecules, with e.g. partly molecular states, antisymmetric ground state, or molecular states for only one type of charge carriers (either electrons or holes).

QDMs find their application potential as quantum gates for quantum information processing.

Here, individual QDs host qubits – two-state systems which, contrary to ordinary bits, can be in a state of a superposition of 0 and 1. Qubits are most often represented by a spin of a charge carrier confined in a QD, but can be also related to the properties of exciton. Quantum gates then allow to perform operations on qubits. Most prominent operation performed by a quantum gate is CNOT (controlled not or controlled negation). It takes two qubits as its input (control and target qubit) and reproduces or flips the target qubit on output depending on the state of the control qubit (the negation is performed when the state of the control bit is 1). CNOT is a universal gate (similarly to NAND and NOR gates in classical digital electronics) – any logical operation can be realized by combining CNOT gates. Various realizations of quantum gates relying on QDMs have been proposed [64, 76]. In addition, all-optical scheme for the initialization and readout of a spin qubit was realized with a QDM [77]. Outside the field of quantum information properties, QDMs (or double quantum dots) are studied for their unusual transport properties [78].

We have contributed to the field of QDMs by two distinct studies aimed at entanglement in lateral QDMs [63] (Sec. 4.1) and active tuning of the tunnel coupling strength in a QDM [49] (Sec. 4.2).

4.1 Entanglement in lateral quantum dot molecules

In this work we further elaborated Bayer's proposal of qubit formed by a charge carrier in a quantum dot molecule, with the carrier in a top/bottom QD representing 0/1. According to the proposal, a QDM can host two qubits (represented by an electron and a hole), for which an entangled state can be formed. Two drawbacks of the proposal limits the entanglement: (1) The QDs in a vertical arrangement are always dissimilar. (2) The tunneling rate for electrons and holes is significantly different.

We studied theoretically a lateral QDM, in which both drawback are resolved. (1) The QDs in a lateral arrangement can be in principal identical. (2) The lateral tunneling rates for both types of charge carriers are not so different as the vertical ones. First, we employed a simple model (derived from a two-site Hubbard model) to study the effect of relevant parameters: The tunneling rates of electrons and holes t_E and t_H, the Coulomb coupling energy U, and the detuning Δ (a difference of the exciton energies in the left and right QD). We have come to the conclusion that for a large degree of the entanglement, $\Delta \ll t_E \approx t_H \ll U$ is required. The antisymmetric direct exciton offers a larger degree of entanglement than the symmetric direct exciton.

Next, we addressed realistic QDMs. For several distances between the QDs forming the QDM, the ratio t_E/t_H varied between 2 and 3, i.e., it was considerably smaller than in vertical QDMs. The entropy of entanglement $S = 0.99$ was predicted for the optimum case, but it was reduced to $S = 0.20$ even for a tiny difference between the QD radii of 1 % (0.2 nm of 14 nm). We proposed a strategy to increase the accessible entropy of entanglement consisting of the following steps: (1) Reducing the distance between the QDs in a QDM. (2) Reducing the lateral dimension of QDs and improving the barrier properties. (3) Minimization of the detuning by improving the homogeneity of QDs.

4.2 Active tuning of coherent tunneling in quantum dot molecules

In this work we demonstrate the possibility to manipulate the coupling strength in a QDM by using externally induced strain fields. Theory explains the effect in terms of modified weight of the light hole component mediating the coupling in the barrier.

The device consists of QDMs (two vertically stacked InGaAs QDs) embedded in the intrinsic region of n-i-p nanomembranes integrated onto piezoelectric actuators. It allows strain and electric field to be applied to single QDMs. Micro-photoluminescence measurements recorded while sweeping the external electric field applied to the molecule revealed characteristic anticrossing pat-

terns [79], from which the tunnel coupling strength can be determined. The hole coupling strength can be modified by applying external biaxial strain: For a compressive (tensile) strain t_H decreases (increases). For the strain range between -0.04 % (compressive) and 0.02 % (tensile), t_H in different QDMs varies by 27–76 μeV, which is 15–30 % of the original value.

Our calculations reproduced well the observations and explained the tuning mechanism for t_H. Inspection of the wave functions reveals that, despite the overall heavy-hole character, the hole probability density in the barrier region is dominated by the light-hole component. The heavy-hole component penetrates the barrier region only by exponentially decaying tails at the QD boundaries, leaving negligible probability density in the barrier center. The compressive strain increases the energy of light holes with respect to the heavy holes, leading to a reduction of the light-hole component in the barrier and therefore the tunnel coupling.

The strain-tunability of the hole coupling can be exploited in the control of entanglement between spin-qubits in quantum dots.

5 Plasmonic nanostructures

5.1 Imaging of the plasmon resonances

We have employed EELS to image plasmons in gold crescent-shaped plasmonic nanoantennas [80]. These structures are highly tunable, support two non-degenerate dipolar (i.e., optically active) LSPR and are compatible with infrared communication technology. Further, a particularly large field enhancement is expected close to their sharp tips. For these reasons, we considered them good candidates for plasmon-enhanced photoluminescence.

Metal nanoantennas of crescent shape were prepared by focused ion beam milling of a sputtered gold layer. The thickness of the antennas was 20 nm and the lateral dimensions ranged between several tens and a few hundreds nanometers. Silicon nitride membranes with the thickness of 40 nm were used as the substrate. The antennas were imaged by annular dark-field scanning transmission electron microscopy. EELS employed electrons with the energy of 300 keV and its spatial and spectral resolution was 0.3 nm and 0.2 eV, respectively. Our experimental research was accompanied by the numerical simulations based on the boundary element method (BEM) using a free software MNPBEM toolbox together with an in-house developed extension.

EEL spectra recorder for particular spots reveal several peaks corresponding to distinct LSPR. Maps of the loss probability for the loss energy corresponding to a particular peak reveal the spatial distribution of the plasmon near field. Depending on the size of the antennas we were able to resolve up to five LSPR.

From the fundamental point of view it is interesting to consider a transformation of a disk-shaped antenna into a crescent-shaped antenna, and related symmetry reduction. We have studied such a transformation and demonstrated splitting of the disk's dipolar mode into longitudinal and transverse dipolar modes of the crescent. Finally, we have demonstrated tunability of the LSPR energies by modifying structural parameters of crescents.

In the follow-up study, we addressed disk-shaped plasmonic antennas [81]. We discussed the influence of involved fabrication techniques (electron beam lithography and focused ion beam lithography) on the properties of plasmonic antennas and concluded that the former technique produces antennas of better quality.

6 Summary and Outlook

Benson's proposal [71] represents a major motivation for my research work over the last decade. In 2000, it opened the path towards the generation of polarization-entangled photon pairs and, at the same time, it raised the need for quantum dots with zero the fine structure splitting of the bright exciton pair. The FSS is related to the anisotropic exchange interaction; any effect that elongates the wave function of an electron or a hole forming the exciton results into finite FSS.

In 2008, when I entered the field, we were able to fabricate strain-free (and thus also piezoelectricity-free) AlGaAs quantum dots. We were able to reconstruct their structural parameters with a fair reliability and we understood well their optical properties [29]. Unfortunately, these QDs were strongly elongated and exhibited thus a large FSS [43]. We had at our disposal a powerful tuning technique based on the application of the external strain using in-house developed piezoelectric actuators [47]. It turned out to be very useful in reducing the FSS, although only a small part of QDs exhibited FSS values sufficient for the generation of entangled photon pairs [44]. Next progress was related to the improvements in the fabrication of quantum dots. With an improved growth protocol, it was possible to fabricate symmetric strain-free QDs with naturally low FSS [26]. Together with the new generation of the piezoelectric actuators, that allowed to independently control individual components of the applied strain tensor, the emission of entangled photon pairs has been experimentally demonstrated.

Along with the main research line, we tackled numerous related problems. In addition to the bright exciton pair, we studied also the dark exciton pair, which was made optically active by the strain present near the cleaved edge of the sample [48] or higher charge and orbital excitation, including excitonic complexes and excited states of the exciton [29, 47]. The external strain field turned out to be a very useful tool for tuning and controlling various properties of quantum dots including the binding energies of excitons and their complexes [47], valence band mixing and polarization anisotropy of light emission [45], or tunneling rates in quantum dot molecules [49]. We have also addressed type-II quantum dots with spatially separated electrons and holes [62, 82]. Among others, we predicted the possibility to achieve zero FSS by adjusting the thickness of the capping layer.

Quantum dots determine the properties of excitons, the initial states for the generation of photons. However, as electronic elements, they have no or little influence on the generation process itself, which happens by means of spontaneous emission. Here, optical elements are required that influence the local photonic density of states and allow to control the rate and directionality of the emission. Plasmonic antennas represent an example of such optical elements. They are metallic nanostructures with dimensions comparable to quantum dots. They support localized plasmon resonance that allow to modify the photonic density of states at te nanoscale. When placed to a close vicinity of quantum dots they do not interfere with excitons except for controlling their spontaneous radiative decay. Our aim is to integrate the quantum dots with plasmonic antennas in order to controll the emission of photons. Our preliminary results in this direction include the fabrication of plasmonic antennas and characterization of their localized plasmon resonances using electron energy loss spectroscopy [80, 81].

After the decades of research, the field of semiconductor quantum dots is mature and ready for transfer of technology to industry. Quantum dot lasers are now commercially available [83], and quantum-dot-based single-photon sources or flash memories are expected to follow in the near future. The research in the field will mostly address novel materials, improvement of existing technologies, scalability necessary for mass industry, etc. Fundamentals of the quantum dot are in general well understood. Nevertheless, novel discoveries based on out-of-the-box thinking are still possible [84].

Quantum dots are one of the state-of-the-art light sources for quantum communications, both as single photon emitters and emitters of entangled photon pairs [85]. Our research substantially contributed to the understanding and optimization of the phenomena related to the latter point, in

particular exciton fine structure splitting.

Despite the favourable properties, quantum dots do not represent an ideal source of quantum light for industrial applications. Most importantly, their fabrication is stochastic, which limits the uniformity of individual light sources and hinders their scalability. Next, low operation temperature is required for the emission of quantum light, at least for most widespread arsenide quantum dots. Further issues are related to the complexity of fabrication, difficult extraction of light from a material with the high refractive index, limited tunability, and environmental hazard.

A promising alternative quantum light source emerged recently: Two-dimensional (2D) single-layer semiconductors (hexagonal boron nitride or transition-metals dichalcogenides) exhibit single photon emission at room temperature [85]. The emitters can be prepared on-demand by local elastic strain [86] which offers unprecedented flexibility and controllability. 2D materials are flexible, offer wide-range tunability, and do not suffer from poor light extraction. While their current performance as light sources is in many aspects inferior to quantum dots, there is a substantial prospect for improvement. The field of light emission from 2D materials is fresh and still vastly unexplored. It is also the field into which we will direct our future research effort.

As excellent light sources, 2D materials are promising candidates for applications in the field of nanophotonics. 2D emitters are extremely compact and chemically stable and thus suitable for on-chip integration. Plasmonic antennas operate in deeply subwavelength regime and therefore also allow high-density integration. It is the light itself and its wave length that limits the minimum accessible size of a photonic element. The time has come to harness the near electromagnetic field that beats the diffraction limit, allows for a subwavelength localization and thus open the path towards smaller photonic elements and higher density of integration. In our future research we will focus on integration of quantum light emitters with plasmonic antennas serving for enhanced light emission, steering the direction of the emitted light, or as waveguide couplers.

References

[1] E. Yablonovitch, Phys. Rev. Lett. **58**, 2059 (1987).

[2] S. John, Phys. Rev. Lett. **58**, 2486 (1987).

[3] T. F. Krauss, R. M. De La Rue, and S. Brand, Nature **383**, 699 (1996).

[4] E. M. Purcell, Phys. Rev. **69**, 674 (1946).

[5] H. Y. Ryu and M. Notomi, Opt. Lett. **28**, 2390 (2003).

[6] A. Badolato, K. Hennessy, M. Atatüre, J. Dreiser, E. Hu, P. M. Petroff, and A. Imamoğlu, Science **308**, 1158 (2005).

[7] D. C. Unitt, A. J. Bennett, P. Atkinson, D. A. Ritchie, and A. J. Shields, Phys. Rev. B **72**, 033318 (2005).

[8] D. R. Smith, W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, Phys. Rev. Lett. **84**, 4184 (2000).

[9] W. J. Padilla, D. N. Basov, and D. R. Smith, Mater. Today **9**, 28 (2006).

[10] D. Schurig, J. J. Mock, B. J. Justice, S. A. Cummer, J. B. Pendry, A. F. Starr, and D. R. Smith, Science **314**, 977 (2006).

[11] D. R. Smith, J. B. Pendry, and M. C. K. Wiltshire, Science **305**, 788 (2004).

[12] J. B. Pendry, D. Schurig, and D. R. Smith, Science **312**, 1780 (2006).

[13] H. Walther, B. T. H. Varcoe, B.-G. Englert, and T. Becker, Rep. Prog. Phys. **69**, 1325 (2006).

[14] R. Miller, T. E. Northup, K. M. Birnbaum, A. Boca, A. D. Boozer, and H. J. Kimble, J. Phys. B **38**, S551 (2005).

[15] H. J. Kimble, Phys. Scr. **1998**, 127 (1998).

[16] M. Pfeiffer, K. Lindfors, P. Atkinson, A. Rastelli, O. G. Schmidt, H. Giessen, and M. Lippitz, phys. status solidi (b) **249**, 678 (2012).

[17] J.-S. Huang, V. Callegari, P. Geisler, C. Brüning, J. Kern, J. C. Prangsma, X. Wu, T. Feichtner, J. Ziegler, P. Weinmann, M. Kamp, A. Forchel, P. Biagioni, U. Sennhauser, and B. Hecht, Nat. Commun. **1**, 150 (2010).

[18] B. Frank, P. Kahl, D. Podbiel, G. Spektor, M. Orenstein, L. Fu, T. Weiss, M. Horn-von Hoegen, T. J. Davis, F.-J. Meyer zu Heringdorf, and H. Giessen, Science Advances **3** (2017).

[19] C. R. Kagan, E. Lifshitz, E. H. Sargent, and D. V. Talapin, Science **353** (2016).

[20] N. Ishikawa, M. Sugita, T. Ishikawa, S.-y. Koshihara, and Y. Kaizu, J. Am. Chem. Soc. **125**, 8694 (2003).

[21] J.-Y. Ge, H.-Y. Wang, J. Li, J.-Z. Xie, Y. Song, and J.-L. Zuo, Dalton Trans. **46**, 3353 (2017).

[22] R. S. Wagner and W. C. Ellis, Appl. Phys. Lett. **4**, 89 (1964).

[23] A. F. Raigoza, J. W. Dugger, and L. J. Webb, ACS Appl. Mater. Interfaces **5**, 9249 (2013).

[24] J. H. Davies, *The Physics of Low-dimensional Semiconductors: An Introduction* (Cambridge University Press, 1997).

[25] P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors: Physics and Materials Properties* (Springer, Berlin, 2005).

[26] Y. H. Huo, V. Křápek, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **90**, 041304 (2014).

[27] I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, J. Appl. Phys. **89**, 5815 (2001).

[28] R. Songmuang, S. Kiravittaya, and O. Schmidt, J. Cryst. Growth **249**, 416 (2003).

[29] L. Wang, V. Křápek, F. Ding, F. Horton, A. Schliwa, D. Bimberg, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **80**, 085309 (2009).

[30] M. Sztucki, T. Metzger, V. Chamard, A. Hesse, and V. Holy, J. Appl. Phys. **99** (2006).

[31] J. Stangl, V. Holý, and G. Bauer, Rev. Mod. Phys. **76**, 725 (2004).

[32] T. Maltezopoulos, A. Bolz, C. Meyer, C. Heyn, W. Hansen, M. Morgenstern, and R. Wiesendanger, Phys. Rev. Lett. **91**, 196804 (2003).

[33] M. Morgenstern, N. Freitag, A. Vaid, M. Pratzer, and M. Liebmann, phys. status solidi RRL **10**, 24 (2016).

[34] V. Křápek, P. Klenovský, and T. Šikola, Phys. Rev. B **92**, 195430 (2015).

[35] D. Bimberg, M. Grundmann, and N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, 1999).

[36] S. Maier, *Plasmonics: Fundamentals And Applications* (Springer US, 2007).

[37] A. Kinkhabwala, Z. Yu, S. Fan, Y. Avlasevich, K. Müllen, and W. E. Moerner, Nat. Photonics **3**, 654 (2009).

[38] V. Křápek, K. Kuldová, J. Oswald, A. Hospodková, E. Hulicius, and J. Humlíček, Appl. Phys. Lett. **89**, 153108 (2006).

[39] C. Heyn, A. Stemmann, T. Köppen, C. Strelow, T. Kipp, M. Grave, S. Mendach, and W. Hansen, Appl. Phys. Lett. **94**, 183113 (2009).

[40] S. Sanguinetti, K. Watanabe, T. Tateno, M. Gurioli, P. Werner, M. Wakaki, and N. Koguchi, J. Cryst. Growth **253**, 71 (2003).

[41] A. Rastelli, S. Stufler, A. Schliwa, R. Songmuang, C. Manzano, G. Costantini, K. Kern, A. Zrenner, D. Bimberg, and O. G. Schmidt, Phys. Rev. Lett. **92**, 166104 (2004).

[42] A. Hospodková, V. Křápek, K. Kuldová, J. Humlíček, E. Hulicius, J. Oswald, J. Pangrác, and J. Zeman, Physica E **36**, 106 (2007).

[43] J. D. Plumhof, V. Křápek, L. Wang, A. Schliwa, D. Bimberg, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **81**, 121309 (2010).

[44] J. D. Plumhof, V. Křápek, F. Ding, K. D. Jöns, R. Hafenbrak, P. Klenovský, A. Herklotz, K. Dörr, P. Michler, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **83**, 121302 (2011).

[45] J. D. Plumhof, R. Trotta, V. Křápek, E. Zallo, P. Atkinson, S. Kumar, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **87**, 075311 (2013).

[46] G. E. Pikus and G. L. Bir, Sov. Phys. – Solid State **1**, 1502 (1960).

[47] F. Ding, R. Singh, J. D. Plumhof, T. Zander, V. Křápek, Y. H. Chen, M. Benyoucef, V. Zwiller, K. Dörr, G. Bester, A. Rastelli, and O. G. Schmidt, Phys. Rev. Lett. **104**, 067405 (2010).

[48] Y. H. Huo, V. Křápek, O. G. Schmidt, and A. Rastelli, Phys. Rev. B **95**, 165304 (2017).

[49] E. Zallo, R. Trotta, V. Křápek, Y. H. Huo, P. Atkinson, F. Ding, T. Šikola, A. Rastelli, and O. G. Schmidt, Phys. Rev. B **89**, 241303 (2014).

[50] R. Trotta, P. Atkinson, J. D. Plumhof, E. Zallo, R. O. Rezaev, S. Kumar, S. Baunack, J. R. Schröter, A. Rastelli, and O. G. Schmidt, Adv. Mater. **24**, 2668 (2012).

[51] R. Trotta, E. Zallo, C. Ortix, P. Atkinson, J. D. Plumhof, J. van den Brink, A. Rastelli, and O. G. Schmidt, Phys. Rev. Lett. **109**, 147401 (2012).

[52] M. D. Biegalski, K. Dörr, D. H. Kim, and H. M. Christen, Appl. Phys. Lett. **96**, 151905 (2010).

[53] J. E. Avron, G. Bisker, D. Gershoni, N. H. Lindner, E. A. Meirom, and R. J. Warburton, Phys. Rev. Lett. **100**, 120501 (2008).

[54] Y. Jang, T. Badcock, D. Mowbray, M. Skolnick, J. Park, D. Lee, H. Liu, M. Steer, and M. Hopkinson, Appl. Phys. Lett. **92**, 251905 (2008).

[55] A. Marent, M. Geller, A. Schliwa, D. Feise, K. Pötschke, D. Bimberg, N. Akçay, and N. Öncan, Appl. Phys. Lett. **91**, 242109 (2007).

[56] M. Geller, A. Marent, T. Nowozin, D. Bimberg, N. Akçay, and N. Oncan, Appl. Phys. Lett. **92**, 092108 (2008).

[57] R. Laghumavarapu, A. Moscho, A. Khoshakhlagh, M. El-Emawy, L. Lester, and D. Huffaker, Appl. Phys. Lett. **90**, 173125 (2007).

[58] S. Itzhakov, H. Shen, S. Buhbut, H. Lin, and D. Oron, J. Phys. Chem. C **117**, 22203 (2013).

[59] K. Akahane, N. Yamamoto, and N. Ohtani, Physica E **21**, 295 (2004).

[60] M. Zíková, A. Hospodková, J. Pangrác, J. Oswald, P. Krčil, E. Hulicius, P. Komninou, and J. Kioseoglou, J. Cryst. Growth **414**, 167 (2015).

[61] K. Kuldová, V. Křápek, A. Hospodková, J. Oswald, J. Pangrác, K. Melichar, E. Hulicius, M. Potemski, and J. Humlíček, phys. status solidi (c) **3**, 3811 (2006).

[62] P. Klenovský, V. Křápek, D. Munzar, and J. Humlíček, Appl. Phys. Lett. **97**, 203107 (2010).

[63] V. Křápek, P. Klenovský, A. Rastelli, O. G. Schmidt, and D. Munzar, J. Phys. Conf. Ser. **245**, 012027 (2010).

[64] G. Burkard, D. Loss, and D. P. DiVincenzo, Phys. Rev. B **59**, 2070 (1999).

[65] H. A. Kramers, Proc. Amsterdam Acad. **33**, 959 (1930).

[66] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, and F. Schäfer, Phys. Rev. B **65**, 195315 (2002).

[67] J. J. Finley, D. J. Mowbray, M. S. Skolnick, A. D. Ashmore, C. Baker, A. F. G. Monte, and M. Hopkinson, Phys. Rev. B **66**, 153316 (2002).

[68] T. Takagahara, Phys. Rev. B **62**, 16840 (2000).

[69] M. Kroutvar, Y. Ducommun, D. Heiss, M. Bichler, D. Schuh, G. Abstreiter, and J. J. Finley, Nature **432**, 81 (2004).

[70] T. Takagahara, Phys. Rev. B **60**, 2638 (1999).

[71] O. Benson, C. Santori, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. **84**, 2513 (2000).

[72] V. Křápek, P. Klenovský, and T. Šikola, Acta Phys. Pol. A **129**, A (2016).

[73] S. Lüker, T. Kuhn, and D. E. Reiter, Phys. Rev. B **92**, 201305 (2015).

[74] M. Korkusinski and P. Hawrylak, Phys. Rev. B **87**, 115310 (2013).

[75] J. M. Luttinger and W. Kohn, Phys. Rev. **97**, 869 (1955).

[76] M. Bayer, P. Hawrylak, K. Hinzer, S. Fafard, M. Korkusinski, Z. R. Wasilewski, O. Stern, and A. Forchel, Science **291**, 451 (2001).

[77] D. Kim, S. E. Economou, S. C. Bădescu, M. Scheibner, A. S. Bracker, M. Bashkansky, T. L. Reinecke, and D. Gammon, Phys. Rev. Lett. **101**, 236804 (2008).

[78] R. Hornberger, S. Koller, G. Begemann, A. Donarini, and M. Grifoni, Phys. Rev. B **77**, 245313 (2008).

[79] E. A. Stinaff, M. Scheibner, A. S. Bracker, I. V. Ponomarev, V. L. Korenev, M. E. Ware, M. F. Doty, T. L. Reinecke, and D. Gammon, Science **311**, 636 (2006).

[80] V. Křápek, A. L. Koh, L. Břínek, M. Hrtoň, O. Tomanec, R. Kalousek, S. A. Maier, and T. Šikola, Opt. Express **23**, 11855 (2015).

[81] M. Horák, K. Bukvišová, V. Švarc, J. Jaskowiec, V. Křápek, and T. Šikola, Sci. Rep. **8**, 9640 (2018).

[82] P. Klenovský, M. Brehm, V. Křápek, E. Lausecker, D. Munzar, F. Hackl, H. Steiner, T. Fromherz, G. Bauer, and J. Humlíček, Phys. Rev. B **86**, 115305 (2012).

[83] A. Sandhu and M. Sugawara, Nature Photonics **1**, 392 EP (2007).

[84] X. Yuan, F. Weyhausen-Brinkmann, J. Martín-Sánchez, G. Piredda, V. Krápek, Y. Huo, H. Huang, C. Schimpf, O. G. Schmidt, J. Edlinger, G. Bester, R. Trotta, and A. Rastelli, Nature Communications **9**, 3058 (2018).

[85] I. Aharonovich, D. Englund, and M. Toth, Nature Photonics **10**, 631 (2016).

[86] A. Branny, S. Kumar, R. Proux, and B. D. Gerardot, Nature Communications **8**, 15053 (2017).

Abstract

The thesis is focused on some interesting aspects of semiconductor quantum dots and plasmonic antennas.

Excitonic fine structure splitting in quantum dots is closely related to the generation of polarization-entangled photon pairs for quantum information processing. I explain the relation of the fine structure to the symmetry of the wave function and related properties of quantum dots: morphology, strain field, and piezoelectric field. I discuss a possibility to tune the splitting by external strain field. Close-to-zero splitting required for the entangled photon pairs is experimentally demonstrated for ultrasymmetric quantum dots and quantum dots under external strain field, and predicted theoretically for quantum dots with the type-II confinement.

External strain can be used to engineer also other properties of quantum dots. Some of them are covered in the thesis, including the binding energies of excitons and their complexes, valence band mixing, polarization anisotropy of light emission, or tunneling rates in quantum dot molecules.

To complete the description of the exciton structure in quantum dots, I report the properties of the dark exciton pair, which does not exhibit linear optical response under standard conditions, but can be brightened near the cleaved edge of the sample.

Emission of light from quantum dots can be controlled and enhanced by plasmonic antennas – metallic nanostructures supporting localized plasmon resonance whose near field overlaps with the quantum dot. Within the thesis I report a study of the plasmon resonances in specific plasmonic antennas (gold crescents and discs) using electron energy loss spectroscopy.

Souhrn

Habilitační práce se zabývá vybranými aspekty polovodičových kvantových teček a plazmonických antén.

Rozštěpení jemné stuktury excitonu v kvantových tečkách je úzce spjato s generací polarizačně propletených dvojic fotonů pro kvantové zpracování informace. Vysvětluji souvislost jemné struktury se symetrií vlnové funkce a souvisejícími vlastnostmi kvantových teček: morfologií, elastickým polem a piezoelektrickým polem. Diskutuji možnost ladit rozštěpení vnějším elastickýn polem. Velmi malé hodnoty rozštěpení, potřebné k vytvoření propletených dvojic fotonů, jsou experimentálně demonstrovány pro ultrasymetrické kvantové tečky a kvantové tečky ve vnějším elastickém poli, a dále teoreticky předpovězeny pro kvantové tečky s uvězněním typu II.

Vnější elastické pole může být využito rovněž k ladění dalších vlastností kvantových teček. Některé z nich jsou studovány v této práci, včetně vazebných energií excitonů a jejich komplexů, míchání valenčních pásů, polarizační anizotropie emisních charakteristik, nebo tunelovací síly v molekulách kvantových teček.

Pro úplnost popisu excitonové struktury v kvantových tečkách doplňuji vlastnosti tmavé excitonové dvojice, která za standardních podmínek nevykazuje lineární optickou odezvu, ale může být zesvětlena v oblasti hrany vzorku.

Emise světla z kvantových teček může být řízena a zesilována pomocí plazmonických antén – kovových nanostruktur s lokalizovanými plazmonovými rezonancemi, jejichž blízké pole se prostorově překrývá s kvantovou tečkou. V práci je zahrnuta studie plazmonových rezonancí v různých plazmonických anténách prostřednictvím spektroskopie energiových ztrát elektronů.